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Abstract

Spin injection into GaAs and Si (both n and p-type) semiconductors using Fe_3O_4 is achieved with and without a tunnel barrier (MgO) via three-terminal electrical Hanle measurement. Interestingly, the magnitude of spin accumulation voltage (ΔV) in semiconductor is found to be associated with a drastic increment in ΔV in Fe_3O_4 based devices for temperature <120 K (T_V, the Verwey transition). Such an enhancement of ΔV is absent in the devices with Fe as spin source. Further, the overall device resistance has no drastic difference at T_V. This renders a direct proof that the observed ΔV is not influenced by the so-called metal-to-insulator transition of Fe_3O_4 at T_V. Observations from our elaborate investigations show that spin polarization of Fe_3O_4 has an explicit influence on the enhanced spin injection. It is argued that the theoretical prediction of half-metallicity of Fe_3O_4 above and below T_V has to be reinvestigated.

Introduction

Electrical spin injection and detection using three-terminal Hanle (3TH) technique in semiconductors have attracted a lot of attention in the past few years owing to the successful demonstration of spin injection at room temperature in various systems [1–6]. In the field of semiconductor spintronics devices, spin injection and detection in various materials find a direct application, for example the proposed Datta-Das transistor [7]. It is important to have high spin injection and detection efficiency in order to obtain a good signal-to-noise ratio in any device. Many groups have adopted different techniques like use of tunnel barriers [1–6] and/or materials with high spin polarization such as Heusler alloys [8] and Fe_3O_4 [9], in an effort to enhance the electrical spin injection efficiency at room temperature in various systems.

Magnitude of spin accumulation in any system depends on factors like (i) type of the injector (ii) spin polarization of the ferromagnet/insulator combination (iii) junction resistance (iv) the uniformity of film at the interface, etc. Choosing a suitable spin injector for an efficient spin injection is one of the essential criteria. Materials like Fe_3O_4 and $La_{2/3}Sr_{1/3}MnO_3$ are promising owing to their high spin polarization when compared to their counterparts, ferromagnetic metals such as Fe, Co, CoFe etc. However, though theoretical calculations indicate half-metallicity in Fe_3O_4 crystal [10, 11], spectroscopic experimental observations always show a spin polarization ranging from 40% to 80% at room temperature depending on the orientation, surface morphology and methods of preparation of Fe_3O_4 [12–14]. Similarly, indirect probing of temperature dependent spin polarization from magnetoresistance measurements reveal maximum spin polarization at Verwey transition ($T_V \sim 120$ K) [15, 16]. Hence, the theoretical prediction of half-metallicity of Fe_3O_4 thin film at room temperature is still under debate.

As the name indicates, 3TH devices consist of 3 terminals for the application of current and the measurement of voltage with one common lead at the center which is the device. Schematic of a typical 3TH measurement for electrical spin injection and detection is as shown in figure 1(a). The magnetic layer at the top (i.e. Fe_3O_4 in this case) acts as a spin source which is magnetized to the saturation magnetization in the plane of the layer. Application of a constant current through the Fe_3O_4 thin film makes sure that sufficient spin-polarized carriers are pumped into the semiconductor where the decay of spin polarized carriers will be measured.

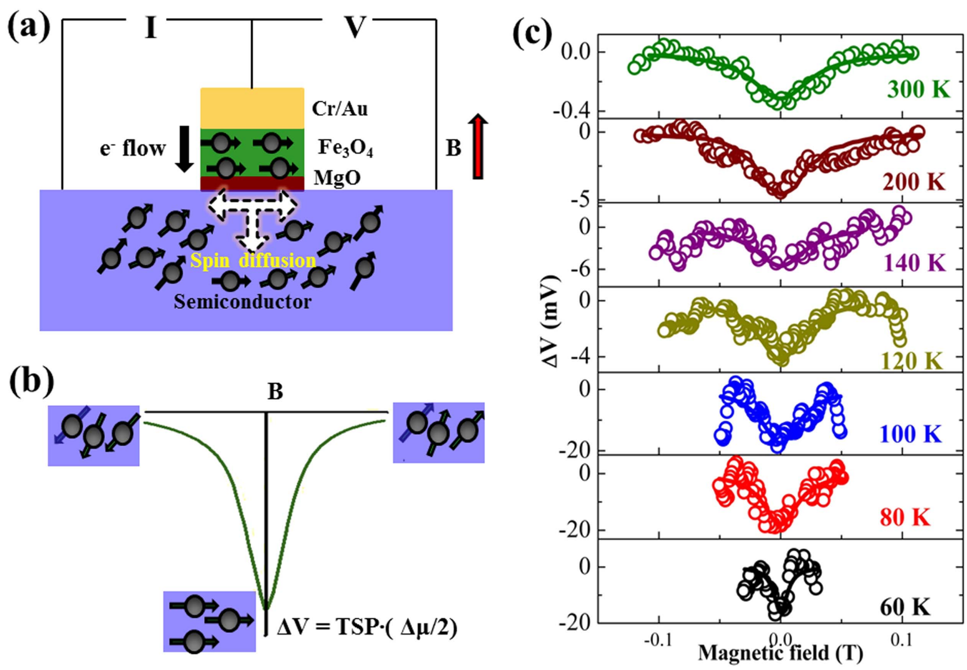

Figure 1. (a) Schematics of the electrical 3TH measurement and the spin relaxation in semiconductor. (b) Schematics of decay of spin accumulation voltage ΔV as a function of perpendicular field B. (c) Plot of temperature dependent measurement of ΔV for a typical 3TH device of Fe_3O_4/MgO/n-type GaAs (10^{18} cm^{-3}) with constant injection current of $I = 1\mu$A. Solid line is the Lorentzian fit for the data at different temperatures.

Insulators are generally used in between metal/semiconductor junctions for better electrical spin injection and detection. In the absence of these tunnel barriers, devices are known to suffer from a conductivity mismatch problem [17, 18]. However, even Schottky barriers are known to be having similar behavior like tunnel barrier [19–21]. Once, the spins are injected into the semiconductor, these accumulated spin-polarized carriers at the junction of semiconductor get scattered or precess depending on the type of semiconductor. To be precise, for example, doping of semiconductor results in the momentum scattering and thus the scattering of spins in the lattice. Whereas, the internal magnetic field due to the spin–orbit coupling from the material results in the precessional motion of spins and thus leads to the spin scattering. Thus, there is a gradient in the measured voltage between the device and the far end of semiconductor due to the spin scattering process within the semiconductor. The accumulated voltage right at the interface of device region with respect to the ground (ΔV) depends on how fast the spin-polarized carriers get relaxed (i.e. spin relaxation time) and how far these carriers can traverse within the semiconductor (i.e. spin diffusion length). In order to scatter the spins in a controlled manner inside the semiconductor to have a better control over the rate of spin relaxation, a perpendicular magnetic field B to the device plane is applied. As the strength of B is increased, spins are forcefully dephased from the initial direction of in-plane, and thus resulting in reduced ΔV. The spin accumulation voltage signal is found to decay as a Lorentzian function of B, $\Delta V(B) = \Delta V(0)/(1 + (\omega_L\tau)^2)$, where $\omega_L = g\mu_B B/\hbar$, is the Larmor frequency of spin precession and τ as effective spin relaxation time. Decay of spin signal ΔV in the semiconductor with perpendicular magnetic field is depicted in figure 1(b). If we look at the magnitude of the accumulation voltage (ΔV) in 3TH geometry of measurement carefully, then ΔV is governed by the factor, $\Delta V = \mathrm{TSP} \times {}^{\Delta\mu}/_{2}$, where TSP is the tunnel spin polarization of the ferromagnet/insulator combination and $\Delta\mu$ the difference in chemical potential for up-spins and down-spins, which is nothing but the spin accumulation at the injected material [1]. Hence, in this regard, spin polarization of the magnetic material together with barrier material (if used) determines the spin accumulation at the interface of the semiconductor. Based on these discussions, we have demonstrated the electrical injection and detection of spins at room temperature in semiconductors like GaAs and Si using Fe_3O_4 with the help of 3TH technique [9, 22].

Methods

In this report, the study of the spin accumulation voltage as a function of temperature is carried out from the experiments based on electrical spin injection and detection. The devices of interest are ranging from an epitaxial injection system Fe_3O_4/MgO/GaAs to polycrystalline Fe_3O_4/MgO/Si junctions with tunnel barriers, including the textured Fe_3O_4/GaAs Schottky junctions. Pulsed laser ablation technique is used to prepare the thin film devices of Fe_3O_4 on GaAs and Si substrates which are deposited under identical temperature of growth (450 °C) and pressure (base vacuum $\sim 10^{-5}$ mbar) with the MgO deposition temperature being 500 °C. Subsequently, device structures are fabricated using photo-lithography with structure details similar to the dimensions discussed in [9]. As mentioned earlier, in any 3-terminal device, the spin accumulation voltage is measured as a function of perpendicular magnetic field B at different temperatures. A typical temperature dependent ΔV versus B of Fe_3O_4/MgO/GaAs(n-type 10^{18} cm^{-3}) device is shown in figure 1(c). As the temperature decreases, the FWHM of the spin signal is reduced due to the lower scattering rate of spins in GaAs. The Lorentzian fit (solid lines in figure 1(c)) to temperature dependent spin signal, ΔV, can be used to extract the spin relaxation time in GaAs depending on the type of doping at a given temperature [9]. An extensive study of the temperature dependence of spin injection and detection in different devices of Fe_3O_4/MgO/Si (both n-type 1×10^{19} cm^{-3} and p-type 3×10^{18} cm^{-3}) and Fe_3O_4/GaAs (n-type 10^{18} cm^{-3}) Schottky systems show a systematic variation of spin-relaxation time for different doping concentrations of substrates [22]. Even in the absence tunnel barrier material, spin injection and detection is shown to be equally possible, as is evident from the earlier studies [21]. However, the interesting observation of the magnitude of spin injection voltage (ΔV) as a function of temperature allowed us to compare various systems at different temperatures. Strikingly, we have found that there is an anomalous increment of ΔV below Verwey transition. Control experiment using Fe/MgO/GaAs devices shows us that the increase in ΔV is unique to Fe_3O_4 based spin injector devices.

Results and discussions

We have investigated the spin accumulation in our devices, like (i) (100) oriented Fe_3O_4/MgO/n-type GaAs, (ii) polycrystalline Fe_3O_4/MgO/n-type Si, (iii) (111) oriented Fe_3O_4/n-type GaAs and (iv) (100) oriented Fe/MgO/n-type GaAs systems (either using metallic or oxide magnetic material as a spin injector into systems like GaAs and Si with and without MgO as a tunnel barrier) [9, 22]. In this report, the magnitude of spin accumulation voltage ΔV in both GaAs as well as Si is studied as a function of temperature with Fe_3O_4 as the spin injector. These observations are then compared with the 3TH voltage obtained from the devices where Fe is used as a spin source. Plot of ΔV as a function of temperature for different injection systems (having same device area of 100×200 μm^2 with a constant injection current of 1 μA for GaAs and 100 μA for a device area of 100×200 μm^2) are given in figure 2.

There is a sharp increase in the spin accumulation at $T < 120$ K for the devices with Fe_3O_4 as a spin injector, which coincides with T_V of Fe_3O_4. $T_V \sim 122$ K [23] is a structural transition in Fe_3O_4 lattice, which changes the magnetization, resistivity, heat transport etc drastically. As seen from figure 2(d), device with Fe as a spin injector has no such type of enhancement of ΔV at around 120 K which otherwise exists in Fe_3O_4 based devices (figures 2(a)–(d)). Normalized spin accumulation voltage is plotted in figure 2(e) for different junctions at various temperatures and the comparison of the magnetization versus temperature is shown in figure 2(f). Shaded area in the figure is the region of the Verwey transition of Fe_3O_4 in different films used, which coincides with the region of enhancement of ΔV. In Fe_3O_4 spin injection system, the increase in the value of ΔV at T_V was found to be between 3.3 to 4 times higher irrespective of the relaxation media (GaAs or Si), type of barrier (tunnel or Schottky barriers), the orientation; (100), (111) and crystallanity of Fe_3O_4 used. Generally, a gradual enhancement of spin accumulation is observed with the increase of barrier resistance in the case of metallic spin injector systems [6, 24, 25]. Even the enhancement in the spin signal from room temperature to 5 K observed for a given barrier thickness is feeble in different cases [2–4, 6, 21, 26].

The magnitude of spin accumulation voltage ΔV differs in various injection systems (figures 2(a)–(d)) though the injection current and device area were kept constant for all GaAs devices. This is purely because of the variation of conditions across the film stack for different devices. In our case, the observed electrical spin accumulation voltage is higher in Fe_3O_4/GaAs junctions (2.2 mV for 1 μA at 300 K), whereas it is least in the case of Fe_3O_4/MgO/Si (10.7 μV for 100 μA at 300 K). ΔV is found to be 0.024 mV at 300 K for Fe/MgO/GaAs device, which is comparatively lower by 2 orders of magnitude when compared with Fe_3O_4/GaAs junctions and 1 order lower than Fe_3O_4/MgO/GaAs devices (0.31 mV for 1 μA at 300 K). Hence, the spin accumulation at various device interfaces is found to be different which is purely dependent on the interface conditions [22].

In our case, resistance of the spin injector Fe_3O_4 is highly temperature dependent and the presence of Schottky or the tunnel barrier always ensures the resistance on either side of the barrier to be irrelevant even at temperatures lower than T_V of Fe_3O_4. Here, we point out that though the resistance change in the continuous layer of Fe_3O_4 is observed, it cannot represent the Fe_3O_4 layer in the reduced device geometry. Hence, we have monitored the 2-probe resistance of the device stack including the substrate instead of Fe_3O_4 layer alone which incorporates the high resistance tunnel junction region as well. Thus, it provides us more confidence in ruling out the extrinsic effects at the junction that may arise. Figure 3 is the plot of the 2-probe voltage of the device

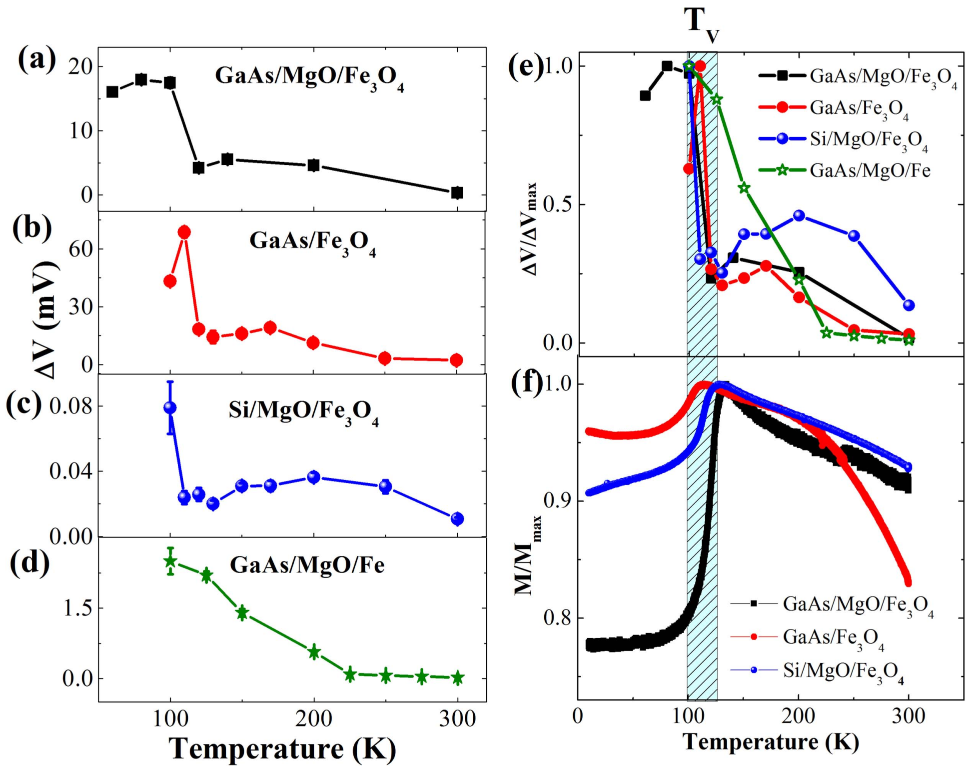

Figure 2. Spin accumulation voltage as a function of temperature for devices of dimension 100 × 200 μm^2 on various systems. Plot of ΔV versus temperature of (a) Fe_3O_4/MgO/GaAs (b) Fe_3O_4/GaAs (c) Fe_3O_4/MgO/Si (d) Fe/MgO/GaAs devices. (e) Comparison of normalized ΔV for all the system. (f) Magnetization versus temperature for Fe_3O_4/MgO/GaAs, Fe_3O_4/GaAs and Fe_3O_4/MgO/Si thin film system showing Verwey transition (shown in shaded region) coinciding exactly the region of enhancement in ΔV as in (e).

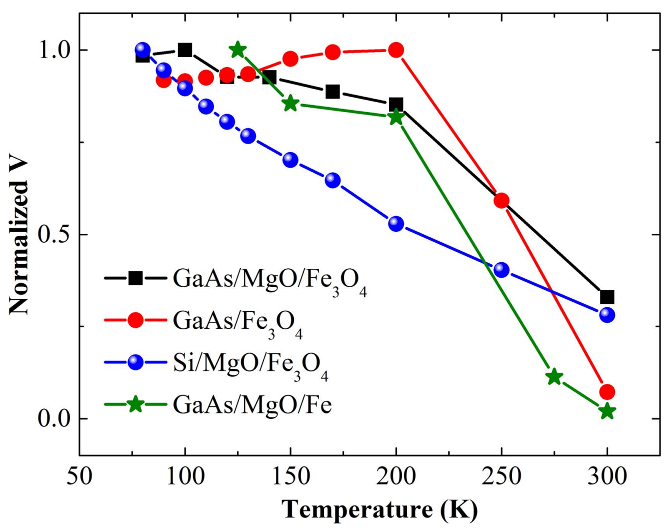

Figure 3. Comparison of normalized value of local 2-probe voltages as a function of temperature for different devices on GaAs for 1 and 100 μA for devices on Si.

stack across T_V. Hence, it can be argued that the enhancement of spin signal is not due to the higher resistance of Fe_3O_4 at low temperatures, unlike the experimental conditions discussed by Wada *et al* [27] using electroluminescence experiment. Since an additional barrier with 2–3 orders of higher resistance is used in our study, we do not observe any effect of the resistance enhancement of Fe_3O_4 layer. The trend in the enhancement of spin polarization observed by Wada and co-workers is gradual across the Verwey transition.

Following the preceding arguments, an anomalous increase in the spin accumulation voltage can be attributed purely to the spin sub-band modification across T_V of Fe_3O_4 but not to the increase in resistance. Since the Verwey transition is associated with a structural modification, configuration of sub-bands of Fe_3O_4

along with the tunnel barrier might influence the total spin accumulation, as ΔV depends directly on the tunnel spin polarization of Fe_3O_4/MgO. But the similar magnitude of enhancement in spin accumulation is even present in the Schottky junction of Fe_3O_4 on GaAs. Hence, the MgO electronic structure bands may not play any vital role in the observed enhanced values of ΔV at lower temperatures.

Enhancement of ΔV in Fe_3O_4 based spin injection could be due to the enhancement of spin polarization in Fe_3O_4 layer. Though Fe_3O_4 is theoretically predicted to be half-metallic in nature [10, 11], experimentally, it has always been a material whose spin-polarization is less than 100% [12, 28, 29] with values varying from 55 to 80%. Fonin *et al* [30] observed that the quality and the growth conditions of thin film highly influence the spin-polarization of Fe_3O_4. For example, spin-polarization of (111) Fe_3O_4 grown on (110) W is found to be −80%, whereas (111) Fe_3O_4 thin film grown on (11–20) Al_2O_3 is only −60% spin-polarization and (001) Fe_3O_4 grown on (001) MgO has spin-polarization of −55% [31]. In all the cases, the spin-polarization of Fe_3O_4 is always higher than that of Fe, which is only 35%. In addition, band structure calculations at lower temperatures also predict the half-metallicity of Fe_3O_4 at $T < T_V$ [32]. But experimental observations by Wang *et al* [16] and Ziese *et al* [15] show that the maximum spin-polarization observed at T_V using magnetoresistance measurements. Even in our case, the behavior of the spin accumulation voltage follows the same trend as discussed by Ziese *et al* [15].

Hence, it is important to mention here that the present band structure calculations of Fe_3O_4 predicts the half-metallicity even at lower temperature ($T < T_V$), i.e., invariance of spin-polarization below and above Verwey transition of Fe_3O_4 [10, 11]. However our experimental results strongly suggest that the enhancement of spin accumulation in Fe_3O_4 based devices is caused by an enhancement of spin-polarization across the Verwey transition. Hence, there is a need to revisit the band structure calculation of Fe_3O_4 in order to solve the long standing puzzle of half-metallicity in Fe_3O_4.

Conclusions

In conclusion, we have demonstrated all electrical spin injection and detection in various systems like GaAs and Si with the help of highly efficient spin injection from an oxide magnetic material Fe_3O_4. Spin accumulation voltage measured in GaAs using 2 different spin injectors like Fe_3O_4 and Fe is compared side-by-side, and the importance of a highly spin-polarized material like Fe_3O_4 as a spin injector is justified. Also, in the absence of a tunnel barrier like MgO, spin injection and detection in Schottky junction of $Fe_3O_4/GaAs$ is observed, in fact, with a higher magnitude of spin accumulation voltage. A huge enhancement in the spin accumulation voltage at $T < 120$ K is observed in the devices with Fe_3O_4 as a spin injector, which is absent in devices with Fe films. From this it can be concluded that the T_V of Fe_3O_4 film has a direct influence on the magnitude of spin accumulation and the resistance enhancement of Fe_3O_4 film itself has no vital role in the observed enhancement of spin accumulation. Moreover, these results are consistent with the variation of spin polarization of Fe_3O_4 with the temperature. Hence, from our study, it can be seen that the spin injection and detection into conventional semiconductors like GaAs and Si can be improved with the help of materials like Fe_3O_4 with higher spin-polarization. Due to enhanced values of the spin accumulation, Fe_3O_4 seems to be promising for the future semiconductor spintronics device applications.

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